MOTION AND THERMODYNAMIC INSTABILITY OF A LIQUID IN A VARIABLE CAPILLARY

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The flow of a liquid in a capillary of variable cross section formed by moving surfaces is analyzed. It is shown that a liquid flow with definite parameters (viscosity, surface tension, etc.) is characterized by a definite negative (relative to atmospheric) pressure, which is responsible for thermodynamic instability of the liquid layer and the formation of discontinuities in it. The theoretical results are consistent with the experimental.

One method of wetting the surface of a solid with a thin layer of liquid is to draw a plate at a constant rate V between rollers. This method is used, in particular, to deposit a uniform homogeneous photosensitive solution on a plate with drillholes having a diameter d of order 10^{-1} cm in the manufacture of advanced hardware communication subsystems. As they are drawn between the rollers the plates are wetted with the solution without the latter entering the holes. After evaporation of the solvent the surface of the plate is left with a uniformly deposited photosensitive layer. The theoretical analysis of the flow process involving the liquid layer between a rotating solution-applicator roller and a plate from the standpoint of physicochemical hydrodynamics makes it possible to expose all the characteristic parameters of the process of practical significance, including the velocity distribution of the layer in its cross section, the excess-pressure distribution in the direction of motion of the layer, and the thickness of the layer deposited on the plate. The conditions for thermodynamic stability or instability of the system, on the other hand, are determined by the magnitude and sign of the excess pressure in the layer.

The motion of a liquid between a roller (of radius R and length L much greater than the layer thickness h) rotating at a linear speed V and a plate moving horizontally at the same speed is one example of flow in a capillary of variable cross section, where viscosity plays a dominant part. The flow of liquid through the capillary onto a metal plate in this case takes place under conditions of heavy loading (static clamping pressure to the plate) of the roller, which has a smooth or fluted elastic surface for deposition of the liquid layer with varying degrees of thinness.

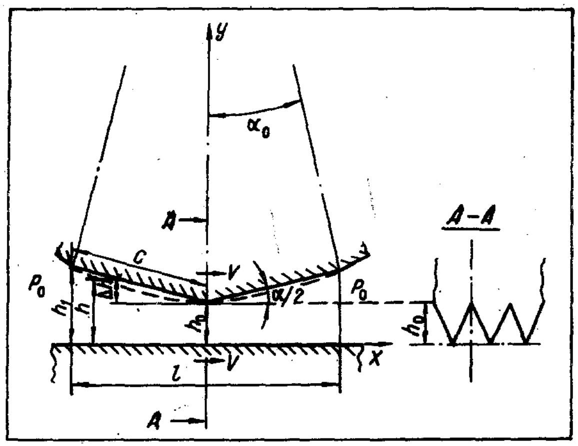

Fig. 1. Capillary of variable cross section.

Three distinct cases of liquid flow are of special interest:

1) flow through identical triangular capillaries with a cross section of variable height in the direction of flow, where the capillaries are formed by fluting grooves on the elastic surface of the roller and by the surface of the plate (Fig. 1);

2) flow through rectangular capillaries of variable height, formed between fluting ridges and the plate surface without a preset gap h_0^x between them at the narrowest part of the capillary; a gap h_0^x and, hence, the possibility of liquid flow through the capillary are realized only when certain conditions are met;

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3) flow through a rectangular capillary of variable height in the direction of flow, where the capillary is formed between the smooth surface of the roller and the plate, again without a preset gap h_0^{xx} between them.

The pressure difference due to the flow of liquid in the direction of the X axis (Fig. 1) through each triangular capillary of height h_0 at its narrowest part is created by the fact that the pressure P developed by constriction of the layer under the roller exceeds the atmospheric pressure p_0 and the additional pressure p under the liquid meniscus in the capillary: $\Delta P = P-(p_0 \pm p) = P-P_0$. The sign of the additional pressure under the meniscus is determined by the wettability of the capillary walls by the liquid. The inception of liquid flow through rectangular capillaries in both of the cases cited above becomes feasible when the normal static pressure P_s applied to the roller (and to the layer) for drawing of the plate equalizes the friction stress ($\tau = \mu\, \partial u/\partial y$) of the liquid against the wall. The normal pressure P_s referred to unit surface is transmitted to any elementary volume designated in the liquid layer. The action of the pressure P_s along the Y axis is equalized by the reaction of the capillary walls, but the action along the X axis is not equalized and causes liquid to flow in that direction, creating and sustaining gaps h_0^x or h_0^{xx} at the narrowest part of the capillaries.

The flow of liquid through rectangular capillaries under the condition $P_s = \mu\partial u/\partial y$, which is necessary for the gaps h_0^x or h_0^{xx} to be "self-sustaining," is activated by the pressure difference ΔP.

Furthermore, in the case of a high wettability of the capillary walls with the liquid the additional pressure under the concave meniscus is negative relative to atmospheric, and liquid flow takes place for a pressure difference $\Delta P = P-(p_0-p)$. In the event of poor wettability of the walls the additional pressure under the meniscus is positive to P_0, the value of $\Delta P = P(P_0 + p)$ is smaller, and it is possible for the liquid not to flow through the capillary. The latter relations are a consequence of well-known tenets of the theory and practical engineering of surface phenomena [1].

We now investigate typical situations of liquid flow through a variable capillary. This problem is reminiscent in part of the motion of a lubricant between rubbing surfaces [2], although it differs in a number of singular respects. Principal among the latter are the flow of liquid in the absence of a preset gap between the roller surface and the plate, as well as the flow of liquid between surfaces made of different materials and moving at the same uniform speed V.

Each of the liquid-flow cases spelled out above is typified by the following general conditions.

The capillary height h varies in the direction of motion of the layer along the X axis according to a linear law (for $R >> h$): $h = \Delta h + h_0 = (a-X)\cdot\alpha/2$, where α is the angle subtending an arc of radius R or, approximately with 3% error or less, the chord of length C, $a = h_1/\tan(\alpha/2)$, and h_0 is the height of the capillary at its narrowest cross section.

The capillary length l is small in comparison with the length of the plate and the circumference of the applicator roller, but it is large in comparison with the height h. The capillary width corresponds to the length L of the applicator roller.

The pressure at the ends of the capillary depends on the external conditions and is determined by P_0. Because of the small layer thickness h the pressure gradient in it does not depend on y and is determined by the pressure gradient $\partial P/\partial X$ in the direction of motion of the layer.

For equal speeds of the applicator roller and plate the relative velocity of the liquid layer at these surfaces is uniquely determined by the speed u_0 of the layer at the plate surface and its speed u_1 at the roller surface and depends on the wettability of those surfaces by the liquid layer.

Corresponding to the motion of the layer and depending on the form of the roller surface (smooth or fluted) are boundary conditions characteristic of the cases described above.

For liquid flow through a capillary formed by fluting grooves and a plate surface the equation of motion with the inertial forces neglected due to the small accelerations involved, has the form

$$\frac{d^2u}{dy^2} = \frac{1}{\mu}\,\frac{dP}{dX} \tag{1}$$

and is subject to the boundary conditions

$$u = u_0 \text{ for } y = 0,\ u = u_1 \text{ for } y = h. \tag{2}$$

For liquid flow through a capillary formed by fluting ridges and a plate surface the equation of motion of the layer corresponds to expression (1) and is subject to the same boundary conditions (2) plus the additional boundary condition

$$P_s = \mu \left(\frac{du}{dy} \right)_{y=h} . \tag{3}$$

The pressure P_s referred to unit surface is uniformly applied along the generatrix of the roller, which consists of n fluting ridges, and it is equal to

$$P_s = \frac{E\Delta R}{R} \cdot \frac{L}{n\Delta x} . \tag{4}$$

The flow of a liquid layer through a variable capillary in the case of a smooth roller surface is also characterized by the equation of motion (1), the main boundary conditions (2), and an additional boundary condition of the type (3).

The pressure P_s^x referred to unit surface is applied to the roller generatrix and is equal to

$$P_s^x = E \frac{\Delta R}{R} . \tag{5}$$

The variation of the layer velocity in its cross section can be found by integrating Eq. (1) with respect to y and assuming dP/dX = const:

$$u = \frac{1}{2\mu} \frac{dP}{dX} (y - h) y + u_0 \left(\frac{h-y}{h} \right) + u_1 \frac{y}{h} . \tag{6}$$

The derivative dP/dX can be evaluated from the condition of a constant volume of solution flowing per unit time across different cross sections of a capillary of variable height on the assumption that the solution is incompressible:

$$Q = \int_0^h u du = (u_0 - u_1) \frac{h}{2} - \frac{dP}{dX} \frac{h^3}{12\mu} .$$

For adjacent cross sections h and h + dh separated by an interval dX we have dQ/dX = 0 and

$$(u_0 - u_1) \frac{dh}{dX} = \frac{d}{dX} \left(\frac{dP}{dX} \frac{h^3}{6\mu} \right),$$

$$\frac{dP}{dX} = 6\mu (u_0 - u_1) \frac{h - h_0}{h} . \tag{7}$$

Hence

$$u = (u_0 - u_1) \left[- \frac{3(h - h_0)}{h^3} (y^2 - yh) - \frac{y}{h} \right] + u_0 . \tag{8}$$

Equation (8) describes the velocity distribution in the capillary cross section.

The expression (8) derived above and the boundary condition (3) make it possible to find the "self-sustaining" height in the narrowest part of rectangular capillaries, where that height is equal to the thickness of the layer deposited on the plate.

To satisfy the boundary condition (3) we must put

$$\mu \left(\frac{du}{dy} \right)_{y=h} = \frac{\mu (u_0 - u_1) 2h_0^x}{h^2} \left(\frac{3}{2} - \frac{h}{h_0^x} \right) = P_s , \tag{9}$$

$$\mu \left(\frac{du}{dy} \right)_{y=h} = \frac{\mu (u_0 - u_1) 2h_0^{xx}}{h^2} \left(\frac{3}{2} - \frac{h}{h_0^{xx}} \right) = P_s^x . \tag{10}$$

Putting $h = h_0^x$ in (9) and $h = h_0^{xx}$ in (10), we obtain

$$h_0^x = \frac{\mu (u_0 - u_1)}{E \frac{\Delta R}{R} \frac{L}{n\Delta x}} , \tag{11}$$

$$h_0^{xx} = \frac{\mu(u_0 - u_1)}{E\frac{\Delta R}{R}}. \qquad (12)$$

It follows from (11) and (12) that the thickness of the layer deposited on the plate has a finite value only under the condition $u_0 >> u_1$. The latter (with the speeds of the roller and plate kept equal) is satisfied only if the capillary walls are made of different materials with different degrees of wetting by the solution, because it is the wettability that determines the sign and magnitude of the additional pressure under the meniscus and varies the mass flow of liquid through the capillary.

In lieu of experimental data on u_1/u_0 and u_0/V an approximate analytical value can be found for the capillary "discharge" rate u_1 for a known value of the maximum excess pressure in the capillary between the roller and the plate:

$$u_1 = \frac{dl}{dt} \cong \frac{h_0^2(P_m - P_0)}{16\mu l}. \qquad (13)$$

Expression (8) enables us to find the variation of the excess pressure in the flow of liquid through the capillary after a series of intermediate transformations and calculations (quantity of liquid flowing through the capillary, pressure gradient, and the integrals in the expression for the latter for a given law of variation of the capillary height):

$$P - P_0 = \frac{6\mu(u_0 - u_1)X(l - X)}{h^2(2a - l)}. \qquad (14)$$

The excess pressure calculated according to (14) for the capillary flow of a solution with a viscosity $\mu = 1.15$ p for $V = 4.4$ cm/sec and $(1 - u_1/V) = 0.6$ is given in Fig. 2. The maximum value of the excess pressure in this case, $P_m - P_0 = 1.23 \cdot 10^6$ dyn/cm^2, occurs in the XY plane where the ratio h/h_0 corresponds to a value of 3/2.

It is reasonable to state that the liquid layer becomes detached from the surface on the applicator roller for $h/h_0 = 3/2$.

Thus, the layer detachment condition is given by the relation $du/dy = 0$ at $y = h$. According to expressions (9) and (10) $du/dy = 0$ (if $u_0 > u_1$) for $h/h_0 = 3/2$.

The excess-pressure distribution in the direction of motion of the layer is symmetrical about the Y axis; in the constricting part of the capillary the excess pressure increases from atmospheric (p_0) to P_m and decreases to the negative value $-P_m$ in the expanding part, whereupon it then equalizes to atmospheric pressure.

The increase of the excess pressure in the constricting part of the capillary ensures that the layer will flow onto the plate.

The reduction of the excess pressure in the expending part of the capillary can induce thermodynamic instability of the system with the formation of a new phase, namely a liquid layer under subatmospheric pressure, which is unstable with respect to the formation of a discontinuity (a "bubble void" at negative pressure).

The result of formation of a discontinuity ("bubble" of critical radius) is equalization of the local negative pressure by extension of the new phase on the surface:

$$-P_m + \frac{2\sigma}{r_{cr}} = 0. \qquad (15)$$

The latter relation enables us to estimate the critical radius of a discontinuity formed in the layer for the above-stated conditions:

$$r_{cr} = \frac{2\sigma}{p_m} = 1.10 \cdot 10^{-4} \text{ cm (for } \sigma \simeq 70.0 \text{ dyn/cm).}$$

In accordance with the theory of thermodynamically unstable systems a liquid layer moving in a capillary formed by the surfaces of a moving roller and plate is unstable with respect to discontinuities greater than $r_{cr} \geq 1.10 \cdot 10^{-4}$ cm.

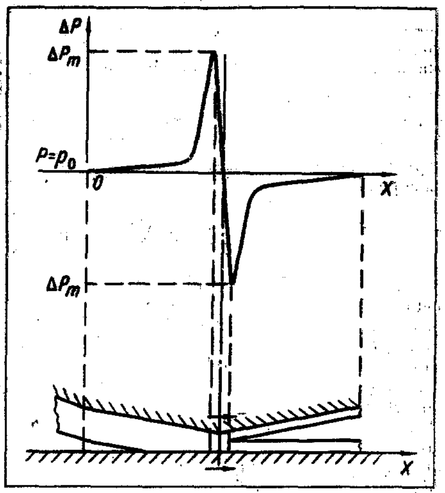

Fig. 2

Condition (15) makes it possible to formulate a criterion for the absence of liquid flow into the mounting holes of a plate, whereby distension of the layer over the holes is possible when the latter have radii smaller than r_{cr}.

For hole radii greater than $1.10 \cdot 10^{-4}$ cm the liquid layer does not distend over the holes and is ruptured at the hole sites. There is no pressure difference along the height of the hole, and the solution cannot flow into them.

The theoretical expressions derived above are in good agreement with experiment. Thus, experiments show that the thickness of a layer deposited on a plate in the roller method is proportional to μ and V and is of the order $2.5 \cdot 10^{-4}$ cm in the dry state (for a viscosity of 1.15 p, $\Delta R/R = 2.10^{-3}$, an elastic modulus $E = 2.8 \cdot 10^{6}$ dyn/cm^2, and a smooth surface on the applicator roller) and that the value calculated according to expression (11) for the thickness of a liquid layer deposited with these parameters and $(1-u_1V) \sim 0.7$ [calculated from Eq. (14) for $u_0 \cong V$] is equal to $6 \cdot 10^{-4}$ cm. Converted to the dry layer this value yields a thickness of order $2.4 \cdot 10^{-4}$ cm.

It is important to add that the maximum excess pressure calculated under the stated conditions for layer flow in a capillary of variable cross section, $P_m - P_0 = 1.23 \cdot 10^6$ dyn/cm^2, agrees in order of magnitude with the additional pressure under a concave surface of a liquid in a cylindrical capillary of diameter h_0: $p = 2\sigma/h_0 \sim 0.6 \cdot 10^6$ dyn/cm^2.

The impermeability of the deposited photosensitive layer into the plate mounting holes of radius $5 \cdot 10^{-2}$ cm $>> r_{cr}$ is also consistent with the results of the experimental work.

The mutual consistency of the analytical and experimental data implies that the method can be reliably used in connection with the analytical parameters of the process.

NOTATION

V	is the speed of the plate and applicator roller forming the capillary walls;
R, l	are the radius and length of the applicator roller;
ΔR	is the required elastic deformation of the roller for drawing of the plate;
E	is the Young's modulus of the elastic surface of the roller;
$\Delta\varkappa$	is the width of the fluting ridges on the elastic roller surface;
n	is the number of fluting ridges on the roller circumference;
h, l, L	are the height of the capillary, its length in the direction of liquid motion, and its width perpendicular to the roller axis;
h_0	is the height and side of equilateral triangular fluting grooves on the roller surface;
$\alpha/2$	is the slope angle of the capillary wall relative to the horizontal axis;
P	is the local pressure of the liquid in the capillary;
P_m	is the maximum value of P;
p_0	is the additional pressure under the liquid meniscus in the capillary;

P_s is the normal static pressure applied to the roller for drawing of the plate;

u is the local velocity of the liquid in the capillary cross section;

u_0, u_1 are the liquid velocities at the surfaces of the plate and roller;

u, σ are the viscosity and surface tension of the liquid in the capillary;

h_0^x is the height at the narrowest part of the capillary formed by fluting ridges on the elastic roller surface and by the plate;

h_0^{xx} is the height at the narrowest part of the capillary formed by a smooth elastic roller surface and by the plate;

r_{cr} is the critical radius of a discontinuity formed in the liquid layer.

LITERATURE CITED

1. Hydrodynamic Theory of Lubrication [in Russian], Moscow (1934).
2. N. K. Adam, The Physics and Chemistry of Surfaces (3rd ed.), Oxford Univ. Press, London (1941).